## Long Range Atmospheric Transport of Radioactive and Chemical Particles -Reflections and Recollections on the 25th Anniversary of the Chernobyl Nuclear Disaster

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Saharan sand storm





Eruption of Mt. St Helen

Industrial pollution

Atmospheric transport of particles is studied in the context of dust storms, volcanic eruptions, atmospheric pollution and nuclear debris released from weapons testing and reactor accidents

Numerical modelling uses methods closely related to those in NWP but some new elements are present...

# **Elements of description**

Frame of reference (Lagrangian or Eulerian)

**Properties of particles**: radioactivity, chemical composition, distribution with sizes, level of toxicity, interaction with clouds and rain

**Meteorological variables:** wind, cloud water, rain fluxes, boundary layer parameters



#### **Transport equations in the Eulerian frame of reference**

$$\frac{d}{dt} \int_{\Omega} \rho dV = -\int_{\Gamma} \rho \mathbf{u} \, d\Gamma \quad \blacksquare \blacksquare$$

$$\frac{\partial \rho}{\partial t} + \nabla \rho \mathbf{u} = 0.$$

Volume  $\Omega$   $e_3$   $e_3$   $e_2$   $e_1$   $e_1$  $e_1$ 

$$\phi = \varphi \, \rho$$

relation between concentration and mixing ratio continuity equation for air

$$\frac{\partial \phi}{\partial t} + \nabla \phi \mathbf{u} = 0.$$

continuity equation for tracer

$$\frac{\partial \varphi \rho}{\partial t} + \nabla \varphi \rho \mathbf{u} = 0 \quad \longrightarrow \quad \frac{\partial \varphi}{\partial t} + \mathbf{u} \nabla f = 0 \quad \longrightarrow \quad \frac{d \varphi}{dt} = 0$$

advection equation for mixing ratio

### The equation is expanded by including molecular diffusion and the source term

$$\frac{\partial \phi_i}{\partial t} + \nabla \mathcal{U}(\mathbf{x}, t) \ \phi_i = \mu \nabla^2 \phi_i + Q(\phi_i, \mathbf{x}, t)$$

The atmospheric flows are genuinely multi-scale



$$\mathcal{U}(\mathbf{x},t) = \mathbf{u}(\mathbf{x},t) + \tilde{\mathbf{u}}(\mathbf{x},t) + \mathbf{u}'(\mathbf{x},t)$$
  $\mathbf{u}(\mathbf{x},t) = \langle \mathcal{U}(\mathbf{x},t) \rangle$ 

The first term on the right hand side is from a meteorological model, terms with tilde and prime denote sub-grid-scale and turbulent fluctuations respectively

After applying the same decomposition to the concentrations of interacting scalar fields we obtain the system which is significantly more complex



# Canonical problem of atmospheric transport with chemical terms

$$\frac{\partial \phi_i}{\partial t} = \underbrace{-\nabla \mathbf{u}(\mathbf{x}, t) \phi_i}_{Advection} - \underbrace{\nabla \langle \mathbf{u}'(\mathbf{x}, t) \phi_i' \rangle}_{Turbulence}$$

$$\underbrace{-\langle \nabla \phi_i \tilde{\mathbf{u}} + \nabla \tilde{\phi}_i \mathbf{u} + \nabla \tilde{\phi}_i \tilde{\mathbf{u}} \rangle}_{Mixing \ Term}$$

$$\mathcal{U}(\mathbf{x},t) = \mathbf{u}(\mathbf{x},t) + \tilde{\mathbf{u}}(\mathbf{x},t) + \mathbf{u}'(\mathbf{x},t)$$

$$\mathbf{u}(\mathbf{x},t) = \langle \mathcal{U}(\mathbf{x},t) \rangle$$

Most of the terms in this system requires some additional "closure" approximation...

Brackets denote averaging, alpha and beta are tensors representing chemical kinetics (typical number of species 50-200)

$$+ \underbrace{\sum_{jk} \alpha_{ijk} \phi_{j} \phi_{k}}_{chemistry} + \underbrace{\sum_{jk} \alpha_{ijk} \langle \phi_{j} \phi_{k} + \phi_{k} \phi_{j} + \phi_{j} \phi_{k} \rangle}_{Subgrid \ scale \ term} + \underbrace{\sum_{jk} \alpha_{ijk} \langle \phi_{j}' \phi_{k}' \rangle}_{Diffusion} + \underbrace{\mu \nabla^{2} \phi_{i}}_{Sources} + \underbrace{\langle \mathcal{Q} \rangle_{i}}_{Sinks} - \underbrace{\langle \mathcal{S} \rangle_{i}}_{Sinks}$$

microscale

The simplest approximation is based on the similarity theory (fluxes are proportional to gradients of resolved quantities, Fick's law)

$$\frac{\partial \phi^{i}}{\partial t} + \nabla \mathbf{u} \, \phi^{i} = \nabla \hat{\mathbf{K}}_{\mathrm{H}} \nabla \phi^{i} + \frac{\partial}{\partial z} K_{zz} \frac{\partial \phi^{i}}{\partial z} + \mathcal{S}^{i}$$

Molecular diffusion is often neglected and the equation is solved with the lower boundary condition expressing the balance of emission and deposition fluxes

The mixing ratio form (known as advection-diffusion equation)

$$\underbrace{\frac{\partial \varphi^{i}}{\partial t} + \mathbf{u} \,\nabla \varphi^{i}}_{\frac{D \varphi^{i}}{D t}} = \frac{1}{\varrho} \nabla \hat{\mathbf{K}}_{\mathrm{H}} \nabla \varrho \varphi^{i} + \frac{1}{\varrho} \frac{\partial}{\partial z} K_{zz} \frac{\partial \varrho \varphi^{i}}{\partial z} + \frac{1}{\varrho} \mathcal{S}^{i}$$

How these simple equations change when we start to consider particles?

The aerosol composed of  $\boldsymbol{m}$  substances can be described by the function

$$n(\mathbf{x}, V_1, \dots V_m) : \mathbb{R}^m \times \Omega \longrightarrow \mathbb{R}$$

Concentration of particles with certain ratios of species inside of the particle measured by partial volumes

 $\sum_p V_p = V$  (*V* is the particle volume), **x** is the coordinate,  $\mathbf{x} \in \Omega \subset \mathbb{R}^3$ 

The actual nuclear release model, see compedium by Williams and Loyalka



Formally the chemical aerosol system is described by the transport equations with coagulation terms introduced by M. Smoluchowski in a seminal 1916 publication, the term K is known as the coagulation kernel.



M. Smoluchowski

## Quick summary of Eulerian frame of reference

- Transport is described by a coupled set of advection-diffusion equations with chemical reaction terms
- The aerosol processes are governed by the stochastic coagulation equation
- closure is based on a similarity theory similar to the one used in the parameterization of the boundary layer



## Alternative descriptions...



Euler

Lagrange

Dirichlet

The alternative is the Lagrangian description (we follow particles from a certain initial configuration).

It is interesting to note that the "Lagrangian" description was first proposed by Euler and the "Eulerian" one by Lagrange; the confusion is due to Dirichlet.





**Euler relation for flow Jacobian** 



## Stretching and folding of material surfaces quickly develops complex shapes...



When the simple initial configuration is placed in the actual meteorological flow

$$\mathcal{U}(\mathbf{x},t) = \mathbf{u}(\mathbf{x},t) + \tilde{\mathbf{u}}(\mathbf{x},t) + \mathbf{u}'(\mathbf{x},t)$$

even without tilde and prime terms we can observe the phenomenon know as chaotic advection. The example on the scale of the Northern Hemisphere is shown in the next slide



Chaotic advection on the hemispheric scale (experiment with real data, the same principle as tracer experiments in oceanography)

In many applications, such as fluid stirring, pollutant dispersal, it is precisely the information inherent in the 'history of individual particles' that is of central interest.

# Interaction between chaotic advection and turbulence



The interaction between chaotic advection and turbulence is not yet explained by a mathematical theory

It is suspected that turbulence leads to homogenization of structures created by the Lagrangian turbulence



Before building the systematic theory a more complete understanding of the kinematics of mixing is required





## Dilemmas of dispersion modelling

Small scale: mainly turbulence (Langevin equations describing random trajectories)

Mesosocale: transition between turbulence and quasi 2D stirring

Large scale: Often the chaotic mixing punctuated by convection

The advection diffusion equation provides good first approximation

## Numerical methods

- Fractional steps methods are used to make problem tractable
- Advection: <u>Semi-Lagrangian</u> or Eulerian (FE, FV, Spectral, Moments...)
- Vertical diffusion: implicit schemes
- Horizontal diffusion: explicit
- Noise control: Shape preserving schemes, FCT, TVD methods...
- Chemistry: QSSA, Gear methods, Runge-Kutta-Rosenbrock schemes
- Aerosol: direct approximation, Modal Aersosol Dynamics



The biggest problem is the noise control (current methods offer multitude of limiting techniques)

## Chernobyl nuclear disaster



Damaged Chernobyl reactor



- The disaster occurred on April 26, 1986 with release of about 15 Mg Curie of radioactivity; (200 times more than Hiroshima bomb)
- Detection of the accident was attributed to Swedish nuclear authorities
- source term was estimated from the solution of an inverse transport problem with some constraints provided from nuclear engineering (constrained optimization problem)



Henri Becquerell



Pierre et Marie Curie

### Units used to describe the source term, concentration in air and deposition

Following Chernobyl accident the amount of release was in Ci (in the following years in SI units Bq).

 $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$ 

 $1 \text{ Bq} = 2.7 \times 10^{-11} \text{ Ci}$ 

I Bq - one disintegration per second

Air concentration was given in Bq/kg or in Bq/ cubic meter, dry deposition in KBq/square meter, wet deposition (same units), occasionally in Bq per volume of precipitated water

## Primary interest in: I-I3I, Cs-I37 and Xe-I33

## Actions following the explosion



clouds seeding to remove radioactivity



- The release of radioactivity from the Chernobyl reactor led to the massive evacuations and contamination of food chain in most of Europe
- The large amount of radioactivity moving in the easterly direction was rained out by the massive cloud seeding operation (the biggest ever)
- The damaged reactor was covered by the mixture of boric acid and sand, the release was virtually stopped after 11 days



The same process occurs in nature



The combined effect of dry deposition and cloud seeding is reflected by the current map of radioactive contamination



## Atmospheric transport models in 1986



$$\frac{\partial \varphi^{i}}{\partial t} + \mathbf{u} \, \nabla \varphi^{i} = \frac{1}{\varrho} \nabla \hat{\mathbf{K}}_{\mathrm{H}} \nabla \varrho \varphi^{i} + \frac{1}{\varrho} \frac{\partial}{\partial z} K_{zz} \frac{\partial \varrho \varphi^{i}}{\partial z} + \frac{1}{\varrho} \mathcal{S}^{i}$$

CANERM CAnadian Nuclear Emergency Response Model

- in 1986 the US ARAC system produced the first prediction of the Chernobyl dispersion
- the second prediction was prepared within several hours in Dorval (first results on April 28, 1986)
- Calculations were performed first on pressure levels and later in the sigma coordinates with the meteorological data from objective analysis (I-131, Cs-137, Xe-131)
- The model used was later known as **CANERM**





The model was predicting the position of a radioactive cloud with good accuracy confirmed in the real time by the aircrafts sampling air radioactivity

Pudykiewicz J. (1989): Simulation of the Chernobyl dispersion with a 3-D hemispheric Tracer Model. **Tellus**, 41B, 391-412.

## **Physical processes considered**



 $dc_a/dt = c_w P$ 

$$c_w = c_a \, \epsilon / m \; \; (Junge \; formula)$$

$$P = \mathcal{C}_0 \left(1 - \exp(-(m/m_r)^2) \, m \right)$$

- Boundary layer mixing (similarity theory)

- Dry deposition scheme based on UK measurements performed following the Windscale nuclear accident (October 1957)

-In cloud scavenging of radioactive particles based on the Junge formula for mass transfer between particles and cloud droplets and Sundqvist relation for the rate of the release of precipitation

$$\Lambda = \beta \Lambda_{cl} = \epsilon C_0 \frac{(U - U_0)}{(U_s - U_0)} [1 - \exp(-(m/m_r)^2)]$$



# The international activities following Chernobyl disaster

Adoption of the Convention on Early Notification of a Nuclear Accident AEA

The joint IAEA/WMO Atmospheric Transport Model Evaluation Study (ATMES) was initiated in November 1986

The Regional Specialized Meteorological Centers (**RSMC**) designated to address meteorological aspects of nuclear emergencies were created under auspices of WMO

Following ATMES CANERM was employed as the operational model at CMC (1991-2009)

Study was arranged by prof.W. Kluge, prof. H van Doop and Dr. G. Graziani

IAEA International Atomic Energy Agency





## The work with CANERM led to some additional developments

Following the conference in Ottawa in 1991 the model was used to estimate the effects of nuclear weapons testing

The adjoint version of the model was used to derive methodology of location of unknown nuclear testing in the context of CTBT (1994-2000)

CTBT: seismic, infrasound, radionuclide

Pudykiewicz J. (1998): Application of Adjoint Tracer Transport Equations for Evaluating source parameters, Atmospheric Environment, vol. 32, no. 17, pp. 3039-3050.

**CTBT Comprehensive Test Ban Treaty** 

## The developments after CANERM

problem is considered in three zones





Transition from 3-D turbulent mixing, mesoscale dynamics, very sharp gradients

**local zone**, Large Eddy Simulation (LES) and smoothed particle hydrodynamics, alternatively stochastic trajectories



flow around structures

**EULAG** 



Large scale advection, high order Eulerian schemes for transport, radionuclide monitoring, inverse problems... atmospheric chemistry





## From CANERM to CHRONOS

The semi-Lagrangian technique used in **EULAG** was combined with the kinetic mechanism of Lurman (1987), meteorology was derived from **MC2** 

Chemistry was solved with the Young and Boris scheme

The model was successful in prediction of high concentration of the tropospheric ozone during smog episodes in the East Coast region



Pudykiewicz et al., 1997: Semi - Lagrangian modelling of tropospheric ozone. *Tellus*, **49B**, 231-248. **The model was later known as CHRONOS.** 

#### **CHRONOS Canadian Hemispheric and Regional Ozone NOx System**

 CHRONOS advection: semi-Lagrangian chemical numerics: Young and Boris scheme Meteorology: GEM

$$rac{\partial arphi_i}{\partial t} = - \mathbf{u} \, 
abla arphi_i$$

$$\frac{\partial \varphi_i}{\partial t} = \frac{1}{\varrho} \frac{\partial}{\partial z} K_{zz} \frac{\partial \varrho \varphi_i}{\partial z} + \frac{1}{\varrho} \mathcal{E}_i^{\nu}$$
$$\frac{\partial \varphi_i}{\partial t} = \frac{1}{\varrho} (\mathcal{Q}_i - \mathcal{L}_i \varrho \varphi_i)$$

$$rac{\partial arphi_i}{\partial t} = -\mathcal{P}_i$$

The model was operational at CMC (2001-2009) Operational 1 day forecast of tropospheric ozone at (18 Z, August 2, 2001)

## **Earth System modelling**



Flammarion engraving, Paris 1888

The search for unknown and uncharted symbolized by the famous Flammarion engraving characterizes the current efforts in modelling of the terrestrial system The idea is to build a comprehensive system describing all elements of the terrestrial system

The massive computing power is required

The scope of processes to be included is still debated. Good discussion was presented by G. Brunet during the internal RPN/ CMC seminar on March 4, 2011 **B. A. Tinsley (2000) INFLUENCE OF SOLAR WIND ON THE GLOBAL ELECTRIC CIRCUIT, AND INFERRED EFFECTS ON CLOUD MICROPHYSICS, TEMPERATURE, AND DYNAMICS IN THE TROPOSPHERE, Space Science Reviews** 



Massive Solar Flare



Solar wind impacting on the Earth magnetic field

- Sun and Earth magnetosphere (space weather)
- Earth electric circuits
- atmospheric particles and clouds and how they are affected by the electric circuits
- atmospheric flow

#### Solar-Terrestrial system and revision of the Bjerknes-Richardson paradigm or the end of a hydraulic analogy



#### The Tsunami wave as seen from the International Space Station

The recent earthquake in Japan is one of the geophysical phenomena which we can't predict.



# Tsunami Wave Formation

#### The general scheme of the nuclear reactor system in Fukushima Daichi



The tsunami wave inflicted serious damage of the nuclear power plant leading to the complex sequence of events which terminated in the **loss of coolant accident**, hydrogen explosion and overheating the tanks with stored spent fuel rods

Zr + 2H<sub>2</sub>0 ->ZrO<sub>2</sub> + 2H<sub>2</sub>

#### Unit 1 und 3

- Hydrogen burn inside the reactor service floor
- Destruction of the steel-frame roof
- Reinforced concrete reactor building seems undamaged
- Spectacular but minor safety relevant







#### Unit 2

- Hydrogen burn inside the reactor building
- Probably damage to the condensation chamber (highly contaminated water)
- Uncontrolled release of gas from the containment
- Release of fission products
- Temporal evacuation of the plant
- High local dose rates on the plant site due to wreckage hinder further recovery work
- No clear information's why Unit 2 behaved differently







## Fukishima from INES 3 to 7

- some information on the source term...
- most of model results
   freely distributed to
   the public all around
   the world





## **Chernobyl INES 7**

-extreme secrecy
-source term evaluated
only from the inverse
problem
model results
distributed only in the
western countries

## Environmental transport of radionuclides

- All environmental compartments should be considered in order to assure accurate dose calculations
- The radioactive water leak from unit 2 requires the use of models simulating transport of radionuclides in the ocean
- The transport in the air is considered first because of the significant speed of dispersion of material by the winds and turbulent mixing



Environmental compartments considered in the prediction of radionuclides transport (air, ocean, soil system, biota, glaciers...)

The release of radioactivity to air and water is still very complex issue and the special nuclear accident models are being used to obtain the exact numbers

The first quantitative information about the long range atmospheric transport was obtained from CTBT monitoring network (additional information about the isotopic ratios)





GFS Analysis for 12Z 22 MAR 2011

300 mb Jet Stream



# Fukushima cloud as seen by the monitoring network of CTBT



Stations detecting radioactivity are marked with yellow

The data from CTBT permitted correct estimate of the release (from 10% to 50 % of Chernobyl release)

## Very large number of different models were applied



#### Austrian Meteorological Service



**NILU Norway** 

Cs-137 Bq/m<sup>3</sup>



#### EURAD

NOAA HYSPLIT MODEL Forward trajectories starting at 0000 UTC 28 Mar 11 00 UTC 28 Mar GFSG Forecast Initialization



HYSPLIT NOAA US

## Numerical simulations performed at the Canadian Meteorological Centre

The extensive modelling was reported by the Environmental Emergency Response Section of CMC. The team has been very actively involved in performing large and small scale simulations to support the Government of Canada efforts in the emerging nuclear crisis. This was done in close collaboration with the Radiation Protection Bureau (Health and Welfare Canada) and with CNSC (Canadian Nuclear Safery Comission). The team at CMC was also cooperating with CTBT in providing inverse modeling assessments.





# Example of a long-range simulation performed by CMC



Simulation is based on 12 March 2011 data. Model was used to determine when any radiation might arrive in Canada. The large scale modeling was supplemented by the fine scale model runs shown in the next slide

Simulation shows relative low-level air radioactivity in Bq/m<sup>3</sup> from 2011-03-12 at 1200 UTC to 2011-03-19 at 0600 UTC.

## Example of a high-resolution simulation

Automatic runs simulating "what if" release scenarios.

Animation shows relative low-level air radioactivity in Bq/m<sup>3</sup> from 2011-03-24 at 0000 UTC to 2011-03-27 at 0000 UTC.



Cs-137 Bq/m<sup>3</sup>

VISAQ

29.03.2011 Daily Mean



Level 1





Level 1

The daily average concentrations of radioactivity of Cs-137 simulated by EURAD

(G. Ebel, University of Cologne)

EURAD is quite similar in the general layout to CANERM



The source term is about 10% of Chernobyl

VISAQ

## Dry deposition of Cs-137 simulated by EURAD

DD Cs-137 Bq/m<sup>2</sup>

12.04.2011 24 UTC (F+72)

DD Cs-137 Bq/m<sup>2</sup>

03.05.2011 24 UTC (F+72)



# The dry deposition pattern is still debated, according to some sources the maps shown above are underestimated, however, measurements support the values shown

(it is assumed the the source estimated will be available from a system of accident models)

## Wet deposition of Cs-I37 simulated by EURAD

WD Cs-137 Bq/m<sup>2</sup>

12.04.2011 24 UTC (F+72)

WD Cs-137 Bq/m<sup>2</sup>

03.05.2011 24 UTC (F+72)



- The wet deposition is significantly larger
- Multi-compartment environmental models are used for dose evaluation
- Further work on the source term



Radioactivity measured at stations around the world is falling faster than in Japan; explanation - effective height of release is low and material is transported mainly by the local flow in the boundary layer

## The local scale dispersion

- The local scale dispersion models are based on various techniques such as the Particle in Cell, Stochastic Langevin Equation, smoothed particle hydrodynamics...
- Despite that the global radionuclide monitoring network shows the reduction of air activities, the situation in Japan remains very serious according the recent reports of IAEA
- The role of the local scale models will increase in June-August, 2011 with arrival of monsoons



The numerical simulation of the local scale dispersion of radionuclides from Fukushima is continued at CMC in Dorval

Fukushima plant will be most likely surrounded by the structure similar to the one designed by AREVA for the Chernobyl reactor



CASING NEAR REACTOR NUMBER









The damaged reactor was surrounded by steel sarcophagus, the structure is not sound...

Before the final dismantling of reactor the old structure will be surrounded by new sarcophagus (Height 110 m, span 257 m, weight 29 000 tons)

# The emerging techniques for transport modelling

- improved meteorological models
- new techniques for aerosols
- adaptive domain partitioning
- optimum transportation theory (Monge-Kantorovich formalism)
- new and flexible grids





# Thank you for listening

